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Angle-dependent magnetoresistance oscillations and Fermi surface reordering at high magnetic fields in α–$(ET)_2KHg(SCN)_4$

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Abstract

Angle dependent magnetoresistance oscillations (AMRO) have been studied in the charge transfer salt α–$(ET)_2KHg(SCN)_4$ for magnetic fields in the range 0 – 30 T. This salt exhibits the onset of antiferromagnetic order at temperatures T_N ~8–10 K and the presence below this temperature of a region of sharp negative magnetoresistance at a field around 22 T known as the "kink". AMRO have been measured in this salt for a wide range of applied fields since the period, amplitude, and nature of the oscillations can be used to directly infer the character of the Fermi surface (FS) as a function of field. The data indicate that a profound change in the band structure occurs at this kink transition; the high field phase is characterised by quasi-2D oscillations from a closed cylindrical FS which is elongated in the c direction; the low field phase appears to be a spin density wave groundstate, with a FS consisting of a sheet (which is quasi-1D in character and tilted at an angle of ~21° to the b^*c plane) and small closed 2D pockets. It is suggested that the breakdown orbits between the pockets and the 1D sheets are able to account for the various Shubnikov–de Haas frequencies observed below the kink.

1. Introduction

Charge-transfer salts of the form ET_2X (where ET is bis(ethylenedithio)tetrathiafulvalene and X is a monovalent anion) have been the subject of intense experimental study since high quality single crystals became available.[1,2] The ET molecules are linked to each other by overlap of their molecular π-orbitals and stack along side one another, separated by sheets of the anion X, to form a two-dimensional (2D) conductive network. Within this family of materials α-$ET_2MHg(SCN)_4$, (M=K, Tl, Rb or NH_4) were synthesised as modifications of κ-$ET_2Cu(SCN)_2$ in an attempt to obtain a higher superconducting transition temperature (T_c).

The salts are isostructural (the so-called α-phase) and as a consequence have almost identical predicted Fermi surfaces (FS) consisting of a 2D closed hole pocket and a pair of 1D planar FS sheets. The salt with M=NH_4 is a superconductor, with $T_c \sim 1\,K$ The salts with M=K, Tl and Rb remain metallic down to < 100 mK[3] and all show the onset of antiferromagnetic order at temperatures T_N ~8–10 K with the easy axis in the highly conducting ac-plane.

An interesting transition has been observed in the magnetic field dependence of the low temperature resistance of the salts with M=K and Tl, but not with M=NH_4; the resistance increases with magnetic field up to ~ 10 T (at 0.5 K), but then decreases with a region of very sharp *negative* magnetoresistance at ~ 23 T (known as the 'kink'). Above this field, Shubnikov de Haas oscillations are observed superimposed upon a monotonically increasing resistance (in contrast, the NH_4 salt exhibits this behaviour at all fields, not just above 23 T). In the K salt, the magnetoresistance exhibits significant hysteresis below the kink[4–7], particularly when the ac-plane of the sample is tilted with respect to the magnetic field[2].

It has been suggested that this kink transition is the point at which the external field destroys the low temperature antiferromagnetic state[5], but the low temperature bandstructure and the field-induced changes giving rise to the kink have remained the subjects of speculation.

2. AMRO experiments

We have measured angle-dependent MR oscillations (AMRO) in single crystals of α-$ET_2KHg(SCN)_4$ in magnetic fields up to 30 T in order to provide information about the FS topology below and above the kink. Magnetic fields up to 30 T were provided by the Nijmegen Hybrid II magnet. Standard 4-wire AC techniques (5–150 Hz) were used for all measurements, with the current directed in the inter-plane b* direction.[2]

The AMRO at 15 T (Fig. 1) indicate that the low magnetic field bandstructure is dominated by a 1D open

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section of FS inclined at ~21° to the crystallographic b^*c plane; qualitatively similar behaviour has also recently been observed in α-$ET_2TlHg(SCN)_4$[7].

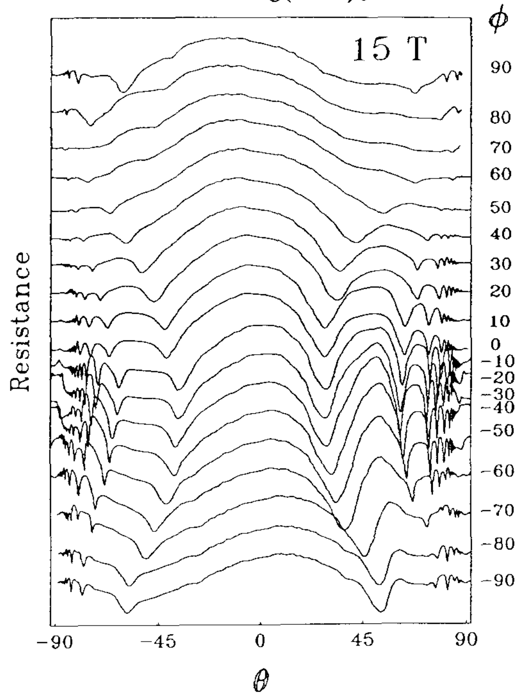

FIG. 1. AMRO of an α-$ET_2KHg(SCN)_4$ crystal at 1.5 K at 15 T as a function of angle θ and ϕ. The traces are offset for clarity.

On raising the magnetic field through the kink, however, the AMRO change in character (Fig. 2), indicating that the FS now posseses a 2D closed section in the form of a distorted cylinder.

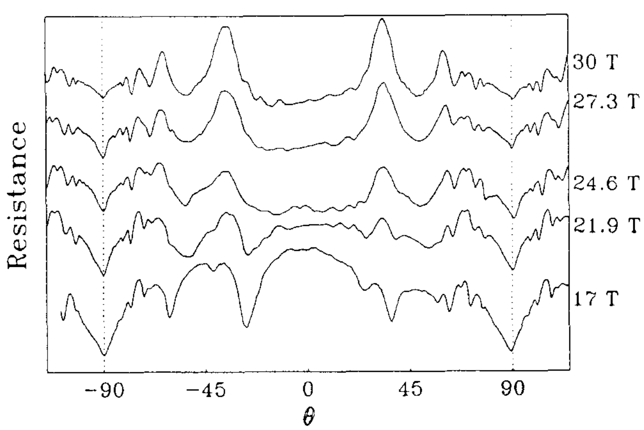

FIG. 2. AMRO in α-$(ET)_2KHg(SCN)_4$ at 1.5 K. The sharp dips seen at 17 T are seen at all lower fields, but change in character in the range 22–25 T.

The action of the proposed SDW state[2] on the calculated bandstructure results in a warped quasi-1D FS tilted by 26° with respect to the b^*c plane, plus small 2D pockets. Above the kink, the magnetoresistance is dominated by 2D orbits indicating a closed cylindrical FS. This transition is seen particularly clearly in Fig. 2 which shows AMRO at fixed ϕ: as the applied magnetic field is increased through the kink transition, dips in the resistance disappear and new peaks appear. This is because the two effects are different in origin: the minima at low field are due to a resonance/commensurability effect of electrons moving across the weakly corrugated 1D sheets[8]; the peaks in the high field AMRO result from the vanishing of the electronic group velocity perpendicular to the 2D layers and are connected with the corrugation of the quasi-2D cylinders.[9]

Acknowledgments

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[1] Organic Superconductors, by T. Ishiguro and K. Yamaji, (Springer-Velag, Berlin 1990); Organic Superconductivity, eds V.Z. Kresin and W.A. Little, (Plenum Press, New York 1990).

[2] J. Caulfield, J. Singleton, P.T.J. Hendriks, J.A.A.J. Perenboom, F.L. Pratt, M. Doporto, W. Hayes, M. Kurmoo and P. Day, *J. Phys.: Condens. Matter* **6**, L155 (1994) and references therein.

[3] H. Ito, H. Kaneko, T. Ishiguro, H. Ishimoto, K. Kono, S. Horiuchi, T. Komatsu and G. Saito, *Solid State Commun.* **85**, 1005 (1993).

[4] J. S. Brooks, C. C. Agosta, S. J. Klepper, M. Tokomoto, N. Kinoshita, H. Anzai, S. Uji, H. Aoki, A. S. Perel, G. J. Athas and D. A. Howe, *Phys. Rev. Lett.* **69**, 156 (1992).

[5] F. L. Pratt, J. Singleton, M. Doporto, A. J. Fisher, T. J. B. M. Janssen, J. A. A. J. Perenboom, M. Kurmoo and P. Day, *Phys. Rev. B* **45**, 13904 (1992).

[6] T. Sasaki and N. Toyota, Solid State Commun. 82, 447 (1992); T. Sasaki, H. Sato and N. Toyota Synth. Met. 42, 2211 (1991).

[7] M. V. Kartsovnik, A. E. Kovalev and N. D. Kushch, *J. Phys. I France* **3**, 1187 (1993).

[8] T. Osada, S. Kagoshima, and N. Miura, *Phys. Rev. B* **46**, 1812 (1992).

[9] M. V. Kartsovnik, V. N. Laukhin, S. I. Pesotskii, I. F. Schegolev, and V. M. Yakovenko, *J. Phys. France* **2**, 89 (1991).